Document made available under the Patent Cooperation Treaty (PCT)

International application number: PCT/CA05/000188

International filing date: 11 February 2005 (11.02.2005)

Document type: Certified copy of priority document

Document details: Country/Office: CA
Number: 2,457,609
Filing date: 13 February 2004 (13.02.2004)

Date of receipt at the International Bureau: 06 April 2005 (06.04.2005)

Remark: Priority document submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b)

PCT/CA 2005/000188
14 MARCH 2005 14.03.05

Office de la propriété intellectuelle du Canada

Un organisme d'Industrie Canada

Canadian Intellectual Property Office

An Agency of Industry Canada

Bureau canadien des brevets

Certification

La présente atteste que les documents ci-joints, dont la liste figure ci-dessous, sont des copies authentiques des documents déposés au Bureau des brevets.

Canadian Patent Office

Certification

This is to certify that the documents attached hereto and identified below are true copies of the documents on file in the Patent Office.

Specification and Drawings, as originally filed, with Application for Patent Serial No: **2,457,609**, on February 13, 2004, by **ALBERTA RESEARCH COUNCIL INC.,** assignee of Partho Sarkar, Hongsang Rho, Gary Kovacik, Luis Yamarte and Rong Zheng, for "Heating Solid Oxide Fuel Cell Stack".

Tracy Faulkner

Agent certificateur/Certifying Officer

March 14, 2005

Date

Canada

(CIPO 68)
31-03-04

OPIC CIPO

Abstract

This invention relates to a solid oxide fuel cell system comprising at least one longitudinally extending tubular solid oxide fuel cell and a longitudinally extending heater mounted in thermal proximity to the fuel cell to provide heat to the fuel cell during start up and during operation as needed. The heater and fuel cell can be encased within a tubular thermal casing; the inside of the casing defines a first reactant chamber for containing a first reactant, such as oxidant. The fuel cell comprises a ceramic solid state electrolyte layer and inner and outer electrode layers concentrically arranged around and sandwiching the electrolyte layer. The outer electrode layer is fluidly communicable with the first reactant, and the inner electrode layer is fluidly isolated from the first reactant and fluidly communicable with a second reactant, such as fuel.

Heating Solid Oxide Fuel Cell Stack

Field of the Invention

This invention relates generally to solid oxide fuel cell (SOFC) systems, and in particular, to thermal management of an SOFC system.

Background of the Invention

In general, an SOFC comprises a pair of electrodes (anode and cathode) that are separated by a ceramic, solid-phase electrolyte. To achieve adequate ionic conductivity in such a ceramic electrolyte, the SOFC operates at an elevated temperature, typically in the order of between about 700°C and 1000 °C. The material in typical SOFC electrolytes is a fully dense (i.e. non-porous) yttria-stabilized zirconia (YSZ) which is an excellent conductor of negatively charged oxygen (oxide) ions at high temperatures. Typical SOFC anodes are made from a porous nickel / zirconia cermet while typical cathodes are made from magnesium doped lanthanum manganate ($LaMnO_3$), or a strontium doped lanthanum manganate (also known as lanthanum strontium manganate (LSM)). In operation, hydrogen or carbon monoxide (CO) in a fuel stream passing over the anode reacts with oxide ions conducted through the electrolyte to produce water and/or CO_2 and electrons. The electrons pass from the anode to outside the fuel cell via an external circuit, through a load on the circuit, and back to the cathode where oxygen from an air stream receives the electrons and is converted into oxide ions which are injected into the electrolyte. The SOFC reactions that occur include:

Anode reaction: $H_2 + O^{=} \rightarrow H_2O + 2e^{-}$

$CO + O^{=} \rightarrow CO_2 + 2e^{-}$

$$CH_4 + 4O^{=} \rightarrow 2H_2O + CO_2 + 8e^{-}$$

Cathode reaction: $O_2 + 4e^{-} \rightarrow 2O^{=}$

Known SOFC designs include planar and tubular fuel cells. Applicant's own PCT application no. PCT/CA01/00634 discloses a method of producing a tubular solid oxide fuel cell by electrophoretic deposition (EPD). The fuel cell comprises multiple concentric layers, namely an inner electrode layer, a middle electrolyte layer, and an outer electrode layer. The inner and outer electrodes may suitably be the anode and cathode respectively, and in such case, fuel may be supplied to the anode by passing through the tube, and air may be supplied to the cathode by passing over the outer surface of the tube. Multiple such fuel cells can be electrically grouped together into stacks to increase power production density.

Because SOFCs can only operate at elevated temperatures, they must be heated before they can generate electricity. During operation, the fuel cells produce electricity and heat. The generated heat can in some instances be used to maintain the fuel cells at their operating temperature; however, in very small scale applications or in other instances, the fuel cells cannot generate enough heat on their own, or there is not enough thermal insulation around the fuel cells to maintain the fuel cells at their operating temperature. In such instances, heat must be provided from an external source. External heating must also be provided at start up, when the fuel cells are not generating any heat.

It is therefore desirable to provide a fuel cell system that can supply sufficient heat to the fuel cells in the system during start up and during operation. In particular, it is desirable to provide a system that can provide such heat in a relatively quick and efficient manner.

Summary of the Invention

According to one aspect of the invention there is provided a solid oxide fuel cell system comprising at least one longitudinally extending tubular solid oxide fuel cell and a longitudinally extending heater mounted in thermal proximity to the fuel cell to provide heat to the fuel cell during start up and during operation as needed. The heater and fuel cell can be encased within a tubular thermal casing; the inside of the casing defines a first reactant chamber for containing a first reactant, such as oxidant. The fuel cell comprises a ceramic solid state electrolyte layer and inner and outer electrode layers concentrically arranged around and sandwiching the electrolyte layer. The outer electrode layer is fluidly communicable with the first reactant, and the inner electrode layer is fluidly isolated from the first reactant and fluidly communicable with a second reactant, such as fuel.

Fuel can be pure hydrogen gas or a reformate produced by a reformer from a hydrocarbon fuel such as natural gas, methanol, or butane. In the case of reformate, the reformer can be mounted inside each fuel cell and comprise a porous reformer catalytic material that allows fuel to flow therethrough and be reformed before reaching the electrochemical reaction area of the fuel cell. Alternatively, the reformer can be mounted inside an extension tube which is mounted to an inlet end of the fuel cell. Alternatively, the reformer can be mounted inside a fuel inlet manifold which is fluidly coupled to the inlet end of one or more fuel cells. Alternatively, the reformer can be mounted in a fuel distribution tube that is inserted inside a single ended tubular fuel cell.

The heater can be an elongate combustor heating tube that combusts fuel in air to produce heat. Fuel can be supplied into the heating tube from a fuel source, or from unreacted exhaust fuel from the fuel cell. The heat produced by the heating tube heats the first reactant in the first reactant chamber which in turn heats the fuel cell by thermal conduction. A catalyst material can be coated on

the inside surface of the heating tube to facilitate catalytic burning of the heating fuel flowing through the heater tube. Alternatively or in addition, the heating tube can be porous to enable fuel to pass from the heater tube and into the first reactant chamber, wherein the heating fuel is combusted to produce heat.

Instead of a combustor heating tube, the heater can be an electric heating element.

In one configuration of the system, a plurality of tubular fuel cells are arranged concentrically around the heating tube. In another configuration, the heating tube is a thermally conductive elongate tube that is large enough to define an oxidant chamber that contains one or more elongate tubular fuel cells. The outside of the heating tube is fluidly communicable with a heating fuel which can be combusted in air to produce heat which thermally conducts into the oxidant chamber and heats the fuel cells therein.

In yet another configuration, the fuel cell system comprises

(a) at least one elongate tubular fuel cell comprising a ceramic solid state electrolyte layer and first and second electrode layers concentrically arranged around and sandwiching the electrolyte layer, the outside of the fuel cell being fluidly communicable with a first reactant fluid and the inside of the fuel cell being fluidly communicable with a second reactant fluid;

(b) a thermally conductive tubular separator surrounding the at least one tubular fuel cell; and

(c) a tubular casing surrounding the tubular separator, the casing being spaced from the separator such that an annular passage is defined therebetween, for receiving a fuel that is burned in the annular passage to produce heat;

wherein some of the heat produced in the annular passage is conducted through the separator to heat the first reactant fluid to a suitable solid oxide fuel cell operating temperature.

Brief Description of Drawings

Figures 1(a) and (b) are schematic top and side sectioned views of a fuel cell system according to a first embodiment of the invention and comprising a plurality of single ended tubular fuel cells.

Figures 2(a) and (b) are schematic top and side sectioned views of a fuel cell system according to a second embodiment of the invention and comprising a plurality of tubular fuel cells open at both ends.

Figure 3 is a schematic sectioned side view of a tubular fuel cell having a reformer reactor installed in a fuel inlet side of the fuel cell.

Figure 4 is a schematic sectioned side view of a reformer reactor installed in an extension tube mounted to a fuel inlet side of a fuel cell.

Figures 5(a) to (d) are schematic sectioned side views showing different embodiments of a reformer reactor installed inside a fuel distribution manifold that is fluidly coupled to a plurality of fuel cells.

Figure 6 is a schematic sectioned side view of a single ended fuel cell and a reformer reactor installed inside a fuel distribution tube fluidly coupled to the fuel cell.

Figure 7 is a schematic end view of a fuel cell stack of a plurality of tubular fuel cells and heating tubes packed within a thermal casing, according to an another embodiment of the invention.

Figures 8(a) and (b) are schematic top and side sectioned views of a fuel cell system according to yet another embodiment of the invention.

Figures 9(a) and (b) are schematic top and side sectioned views of a modified version of the fuel cell system of Figs 8(a) and (b) in which the fuel cells are embedded in a solid state porous foam matrix.

Figures 10(a) and (b) are schematic top and side views of a fuel cell stack separator according to yet another embodiment of the invention.

Figure 11 is a schematic top view of the fuel cell stack separator of Figures 10(a) and (b) installed in a fuel cell system.

Figures 12(a) and (b) are schematic top views of the fuel cell system of Figure 11 with an inner and outer heating tube (Figure 12(a)) and an inner heating tube only (Figure 12(b)).

Figures 13(a) –(d) are schematic top views of fuel cell systems according to other embodiments of the invention, each of the four systems having different combustor designs.

Detailed Description of Embodiments of the Invention

References in this description to directional terms such as "top", "bottom", "side" are used merely for convenient reference when describing the embodiments of the invention, and are not intended to limit the orientation of the embodiments in use or in connection to another component in a system.

When describing the present invention, the following terms have the following meanings, unless indicated otherwise. All terms not defined herein have their common art-recognized meanings.

The term "ceramic" refers to inorganic non-metallic solid materials with a prevalent covalent or ionic bond including, but not limited to metallic oxides (such as oxides of aluminum, silicon, magnesium, zirconium, titanium, chromium, lanthanum, hafnium, yttrium and mixtures thereof) and nonoxide compounds including but not limited to carbides (such as of titanium tungsten, boron, silicon), silicides (such as molybdenum disicilicide), nitrides (such as of boron, aluminum, titanium, silicon) and borides (such as of tungsten, titanium, uranium) and mixtures thereof; spinels, titanates (such as barium titanate, lead titanate, lead zirconium titanates, strontium titanate, iron titanate), ceramic super conductors, zeolites, and ceramic solid ionic conductors (such as yittria stabilized zirconia, beta-alumina and cerates).

The term "cermet" refers to a composite material comprising a ceramic in combination with a metal, typically but not necessarily a sintered metal, and typically exhibiting a high resistance to temperature, corrosion, and abrasion.

Referring to Figure 1 and according to a first embodiment of the invention, a fuel cell system 10 includes a plurality of longitudinally-extending tubular solid oxide fuel cells 12 spaced equally from and around the outside of a longitudinally-extending central heating tube 14 (also known as a "combustor"). The fuel cells 12 and heating tube 14 are surrounded by a longitudinally-extending outer casing 16; the heating tube 14 and the casing 16 define an annular chamber 18 in which the fuel cells 12 reside. The ends of the casing 16 are capped by respective top and bottom end caps 19, which are provided with

openings that serve to hold the fuel cells 12 and heating tube 14 in place and pass oxidant and fuel reactants to and from the system 10 for electrochemically producing electricity ("reactant air" and "reactant fuel"), and for combusting to produce heat ("heating air" and "heating fuel").

The fuel cells 12 are of a micro-tubular type that may be manufactured, for example, by the methods taught in Applicant's published Patent Cooperation Treaty applications PCT/CA01/00634 or PCT/CA03/00059. PCT application PCT/CA01/00634 teaches the production of a tubular SOFC by electrophoretic deposition (EPD) and PCT/CA03/00059 teaches the production of a tubular SOFC by metal electrodeposition (MED) or composite electrodeposition (CED). Micro tubular fuel cells produced by these techniques have a hollow tubular ceramic-containing structure and comprise concentric contacting membrane layers that serve as the anode, electrolyte, and cathode of the fuel cell. In the context of this application, "micro-tubular" SOFC means an SOFC having a diameter of 5mm or less. These micro-tubular fuel cells can have diameters as small as about 10μm, and various cross-sectional geometries, such as circular, square, rectangular, triangular, and polygonal. Although this description primarily describes a fuel cell system using micro-tubular fuel cells with a circular cross-section produced by these techniques, it is within the scope of the invention to use larger diameter tubular fuel cell tubes with non-circular cross-sectional geometries, that are made by other techniques as known in the art.

The inner electrode can be the anode and can have one or more sub-layers. In this embodiment, the inner electrode has three sub-layers (not shown), in which an innermost sub-layer ("1st anode sub-layer") is made by MED or CED and can multiple openings therethrough to allow fuel to reach a middle anode layer (2nd anode sub-layer). The main function of the 1st anode sub-layer is current collection and suitable materials for this sub-layer are a metal such as Ni or Cu (deposited by MED) or a cermet such as Ni (or Cu) and yttria stabilized zirconia or doped ceria (deposited by CED). The 2nd anode sub-layer is

deposited onto the 1st anode sub-layer by EPD and has a composition comprised of a mixture of nickel oxide and yttria stabilized zirconia or doped ceria. This sub-layer also serves to collect current as well as to provide mechanical support for the fuel cell; the sub-layer has a thickness selected to provide suitable mechanical support and thus tends to be the thickest of the three anode sub-layers. A 3rd anode sub-layer is deposited by EPD onto the 2nd anode sub-layer and has a composition comprised of a mixture of nickel oxide and yttria stabilized zirconia or doped ceria. One of the powders must have smaller average particle size than the 2nd anode sub-layer. This 3rd anode sub-layer will have a higher triple phase boundary and the majority of the electrochemical reaction will happen in this sub-layer.

The fuel cells 12 are closed at one end and have an open end that extends above the top edge of the casing 16 and through the top end cap 19. Optionally, the fuel cells 12 can have both ends open as shown in Figures 2(a) and (b). As the inner layer in each fuel cell 12 is the anode layer, the outer layer is the cathode layer. Accordingly, the open end of each fuel cell 12 is coupled to a fuel source (not shown) such that gaseous reactant fuel is transmitted to the inside of each fuel cell 12 for electrochemical reaction. The fuel can be pure hydrogen gas stored in a metal hydride tank, or produced on demand from water by electrolysis, or other methods as is known in the art. Or, the fuel can be a reformate produced by a reformer 102 from a hydrocarbon fuel such as natural gas, methanol, butane etc. The reformer 102 can be integrated into each fuel cell as shown in Figure 3 and Figure 4, or be a separate unit attached to a plurality of fuel cells as shown in Figure 5.

Referring to Figure 3, a tubular fuel cell 12 is open at both ends and has a reformer reactor 102 mounted inside the fuel cell 12 at the fuel inlet side. The reactor 102 comprises reformer catalyst material packed into the fuel inlet side of the fuel cell 12 such that it allows reformation as well as gas flow through. On each side of the catalyst material is a porous stopper 104 that holds the catalyst

material in place within the fuel cell 12. Alternatively, the reactor 102 can comprise a particulate or granular catalyst support structure that is coated with appropriate reformer catalyst material as known in the art. In particular, the reformer 102 can be a ceramic, metal or cermet foam or a porous mass that is coated with catalyst material on the fuel inlet side, or be a porous or foamy anode current collector that is coated with catalyst material.

Referring to Figure 4 and according to another embodiment of the invention, the fuel cell 12 can have an extension tube 106 mounted to the fuel inlet side of the fuel cell 12. The reformer reactor 102 can be totally situated in the extension tube 106 as shown in Figure 4, or partially in the extension tube 106 and partially in the fuel cell (not shown).

Referring to Figures 5(a) – (d) and according to another embodiment of the invention, a fuel inlet manifold 108 is provided that is fluidly coupled to a plurality of fuel cells 12 and has a fuel inlet conduit 110 that receives fuel for distribution to each fuel cell 12. Referring to Figure 5(a), the reformer reactor 102 is situated inside the fuel inlet manifold 108 such that fuel passes through the reactor 102 and to the fuel cells 12. Optionally and referring to Figure 5(b), the fuel reformation pathway through the reactor 102 can be lengthened by installing a fuel inlet guide 116 having a fuel inlet located upstream of the reactor 102, a fuel outlet guide 114 having a fuel outlet located downstream of the reactor 102 and a fuel distributor plate 112 located in a spaced position downstream of the fuel outlet guide 114. Optionally, and referring to Figure 5(c), the fuel inlet guide 116 can be omitted and the fuel reformation pathway length can be maintained by moving the fuel inlet conduit 110 to a lateral position on the manifold 102. Optionally and referring to Figure 5(d), the reactor 102 can be installed in the inlet conduit 110, which enables a longer fuel reformation pathway than in the embodiments shown in Figures 5(a)-(c) using the same amount of reactor material and without using guides 114, 116, since the cross-section of the reactor 102 is reduced.

Though Figures 5(a)-(d) show fuel cells 12 each with both ends open, the fuel cells 12 can be readily adapted to be single-ended. In case of a single ended fuel cell (e.g. as shown in Figure 1), the reformer reactor 102 can be located in the open end of the fuel cell 12 (not shown), or in a fuel distribution tube 21 as shown in the Figure 6. Optionally, in all the above embodiments, the reformer reactor may have more than one reaction zone (not shown) in which each zone has different catalyst material. Also, each zone may have a different operating temperature and the choice of catalyst material for a particular zone can depend on the operating temperature of the zone.

Referring again to Figure 1, the fuel source supplies fuel to the fuel distribution tube 21, which is inserted into each fuel cell 12 such that the reactant fuel is discharged from the distribution tube 21 near the bottom of the fuel cell 12; the reactant fuel then flows upwards and is electrochemically reacted. Excess reactant fuel and reaction products are discharged from the open top end of the fuel cell 12 out of the system 10, or is fed into the heating tube 14 and burned to produce heat for the system 10 (shown as arrow 23), as will be described below.

Reactant air is flowed through an air inlet 25 in the bottom end cap, into the chamber 18, and out of the chamber 18 through an air outlet 27 at the top end cap. The reactant air flows over the cathode surface of each fuel cell 12 and thus provides the oxygen required for electrochemical reaction.

Alternatively, the air inlet can be on the top end cap 19 and the air outlet on the bottom end cap 19, or the air inlet and outlet can be on the same end cap 19. When the air inlet and air outlet are on the same cap 19, a distribution tube (not shown) similar to the fuel distribution tube 21 is connected to the air inlet to flow air from the air inlet to the other end of the fuel cell so that air can flow back over each fuel cell's reaction zone and back to the outlet. Incoming and exhaust air can pass through a heat recuperator (not shown in the figure).

In order for an electrochemical reaction to occur, the fuel cells 12 and their reactants must be at an adequate operating temperature, typically between 500 - 1000°C and particularly around 800°C. Heat is supplied to the fuel cells 12 by combusting fuel in air inside the heating tube 14. The heating tube 14 is made of a thermally conductive material that can withstand typical SOFC operating temperatures, i.e. temperatures up to 1000 °C. Such material includes ceramics such as SiC, Al_2O_3, SiO_2, MgO, and ZrO_2, high temperature metals or metal alloys such as Niconel, stainless steel, ferretic steel, cermets (e.g. a ceramic such as SiC, Al_2O_3 with a metal such as Inconel, stainless steel, ferretic steel, stainless steel), ceramic-coated metals, or metal-coated ceramics. The heating tube 14 is sufficiently porous to allow the flow of air and fuel therethrough and to provide sites for catalyst deposition.

The casing 16 is made from a thermally insulating material such as a ceramic insulator, aerogel, vacuum flask (made from quartz glass, Pyrex glass, stainless steel; when made with glass, the vacuum flask can be covered with a thermally reflective coating such as silver, gold, or any other suitable insulating material as is known in the art) or heat recuperator. The casing 16 can be cylindrical, or have a different cross-sectional geometry. The selected thickness of the casing 16 will depend on the available space in the application in which the system 10 is used; when used in small-scale portable electronic devices, the casing 16 is kept relatively thin for packaging reasons which reduces the effectiveness of the casing 16 to insulate the system 10 from thermal losses. In certain very small applications, the casing 16 is too thin to enable the system 10 from generating enough heat from the electrochemical reaction alone to continuously maintain an adequate operating temperature.

On those occasions where the operating temperature cannot be sustained by electrochemically-produced heat, the heating tube 14 supplies heat from combustion to the system 10 in order to keep the system 10 at a suitable

operating temperature. A pressurized heating fuel stream 29 and an air stream 31 are fed into the heating tube 14 at its top end and flamelessly catalytically burned to produce heat. Unused heating fuel, air and combustion products 33 are exhausted from the heating tube 14 via its bottom end. The pores of the heating tube 14 are coated with a suitable catalytic material such as platinum, palladium or other materials as is known in the art. The product heat warms the reactant air and the fuel cells 12 inside the chamber 18 by radiation and conduction. When supplying heating fuel at a sufficiently high pressure, some of the heating fuel will permeate through the heating tube 14 and combust with reactant air in the chamber 18. The heat released as a result of the combustion will contribute to heating the reactant air in the chamber 18 and the fuel cells 12. The reactant air flow rate through the chamber 18 is managed to ensure that combustion products are removed at a sufficient rate that they do not accumulate inside the chamber 18.

While one row of fuel cells 12 encircles the heating tube 14 in the embodiment shown in Figure 1, additional rows of fuel cells 12 can be provided in the system 10. The number of fuel cells 12 used in the system 10 will depend on part on size restrictions. In particular, in micro electronics or other portable applications, the fuel cell system 10 will have to be kept as small and as light as possible, in which case the system 10 can be configured with fewer fuel cells 12 than in cases where system size is not a limiting factor.

Optionally, the heating tube 14 can be constructed of a dense (i.e. non-porous material) and can be closed at one end. However it is preferable for the heating tube 14 to be porous, as this increases the surface area for catalytic burning, as well as provides the opportunity for combustion to occur inside the chamber 18.

Also optionally, the heating tube 14 can be filled with a solid-state, thermally conductive porous foam matrix (not shown) or other suitable porous

material that is able to withstand SOFC operating conditions, and can be coated with catalytic material to further promote catalytic burning therein.

In operation, the fuel cell system 10 must first be heated to a temperature that will enable the fuel cells 12 to operate; it has been found that when the fuel cells 12 are based on yttria-stabilized zirconia (YSZ) materials, the fuel cells 12 can start to electrochemically produce electricity at about 600°C and when based on doped ceria-based materials, the fuel ells can start to produce electricity at around 450°C. In order to heat the fuel cells 12 to this temperature, the heating tube 14 is used to produce heat on start-up by combusting heating fuel and air.

As the catalytic material must also be heated to an elevated temperature before it can catalytically combust the heating fuel, a burner 35 is provided to ignite the heating fuel to produce sufficient heat to heat the catalytic material to its operating temperature, which is in between about 300-400 °C. The burner 35 is mounted at the upstream (top) end of the heating tube 14 and in the flow path of the heating fuel and air. Optionally, the burner 35 can be mounted in the bottom end of the tube 14 in the flow path of the heating fuel and air. A piezoelectric spark or other suitable sparking means inside the burner is used to ignite the fuel stream 29 passing through the burner 35.

Alternatively, the burner 35 can be replaced by an electric heater (not shown) as is known in art. In particular, a small electric heater is preferably surface mounted to the system 10, and serves to heat a small area to a sufficiently high temperature so that catalytic burning can start at that location and then the catalytic burning can heat up a surrounding area where catalytic burning can expand. In this way catalytic burning will spread throughout the tube wherever catalyst is present.

The system 10 is started by first supplying the pressurized heating fuel stream 29 and air stream 31 through the burner 35 and igniting same to produce

heat. The heating fuel stream 31 can come from the same source as the reactant fuel. The reactant fuel supply to the fuel cells 12 is turned off or optionally can be flowed at a trickle to the fuel cells 12 to purge air or other gases resident in the fuel cells 12. Once the catalyst in the heating tube 14 is warmed to its operating temperature, the heating fuel stream 29 is stopped to quench the flame, then restarted to supply fuel to the heating tube 14 for catalytic burning.

The heat produced by catalytic burning is used to heat the fuel cells 12 to about 450-700°C. Once fuel cells 12 reach this temperature range, they start to produce electricity. The fuel cells 12 then rapidly warm to their ideal operating temperature of about 500-800°C (exact ideal operating temperature depends on the type of electrolyte) and at that time, the heating fuel stream 29 is turned off or reduced. A temperature sensor (not shown) connected to a control system (not shown) is used to monitor the temperature of the system 10; when the temperature falls below a selected lower temperature threshold (i.e. around a temperature where the electrochemical reaction will stop or performance be substantially degraded), the control system turns on or increases the heating fuel stream 29 into the heating tube 14 to produce heat as required to keep the system 10 at its ideal operating temperature. Unreacted fuel 23 from the fuel cells 12 can also be supplied to the heating tube 14, as the electrochemical reaction typically only consumes about 70-80% of the fuel supplied to the fuel cells 12. Control valves (not shown) are provided to control the flow of heating fuel streams 23 and 29 into the heating tube 14.

Alternatively, the heating tube 14 can be supplied fuel entirely from unreacted exhaust fuel 23 (i.e. no separate heating fuel stream 29 is provided). At start up, the fuel cells 12 are not yet producing electrical power and thus the fuel exhaust stream 23 exiting each fuel cell 12 contains approximately 100% fuel (the balance being water vapor etc.). This exhaust fuel stream 23 is fed into the combustors 18 and ignited to produce enough heat to heat up the fuel cell stack. The fuel flow to the fuel cells 12 and to the heating tube 14 can be

controlled so that heat and electricity are both produced in sufficient quantities. Consider for example a fuel cell stack that typically requires 100ml/min of fuel to operate to produce electricity. At start up, an initial fuel flow rate is selected that will be sufficient to operate the heating tube 14 to produce sufficient heat for stack operation; this flow rate may be lower or higher than 100ml/min. As heat is generated by the heating tube 14 and the stack becomes warm enough to produce power, some of the fuel will be utilized by the stack to produce electricity (as well as some heat), and as a result, the amount of fuel in the exhaust fuel stream 23 flowing to the heating tube 14 will decrease. As the stack reaches its operating temperature, less heat is required from the heating tube 14 than at start-up, which conveniently corresponds to a reduced heat production by the heating tube 14 resulting from receiving less fuel from the exhaust fuel flow. Fine tuning of stack temperature can be performed by controlling the air flow rate and fuel flow rate to the fuel cells 12. Combustor and fuel cell exhaust air can pass through a heat recuperator (not shown) that recovers some of the produced heat to be used to heat the stack.

Instead of catalytically burning the heating fuel, the heating fuel may be entirely burned by the burner 23. In such case, no catalytic material is required in the heating tube 14, as the heating fuel stream 31 is ignited by the burner 35 during start up and steady state operation to produce heat for the system 10.

According to another embodiment of the invention, and referring to Figure 7, multiple fuel cells 12 and multiple heating tubes 18 (combustors) are stacked together in an annular chamber 18 inside a thermal casing 16. The combustors 18 are strategically placed amongst the fuel cells 12 in order to provide a uniform distribution of heat to the fuel cell stack. The combustor tube wall can be dense in which case the exhaust fuel flowing through the combustor 18 is catalytically burned inside the combustor 18.

The combustors 18 use unreacted exhaust fuel and oxygen supplied from air distributed throughout the stack to generate heat. In this connection, oxygen supply conduits (not shown) are connected to the air outlet end of the annular chamber 18 and to the inside of each combustor 18, to enable exhaust air to flow to each combustor 18. Alternatively, fresh air can be supplied directly to each combustor 18. Similarly, fuel supply conduits (not shown) are connected to the fuel outlets of each fuel cell 12 and to the inside of each combustor 18 to enable exhaust fuel to flow to the each combustor 18.

According to another embodiment of the invention and referring to Figures 8(a) and (b), a fuel cell system 10 is provided that is similar to the first embodiment, except that in this embodiment, the heating tube 14 is enlarged, and the fuel cells 12 are placed inside the heating tube 14. The space between the heating tube and casing 16 is now defined as the heating chamber 20, and the space inside the heating tube is now defined as the oxidant flow chamber 22. Reactant fuel and oxidant supply and discharge connections are reconfigured such that reactant fuel is supplied to and removed from each fuel cell 12 inside the oxidant flow chamber 22, reactant air is supplied to and removed from the oxidant flow chamber 22, and heating fuel and air is introduced into the heating chamber 20 and combusted to produced heat that is used to heat the oxidant and the fuel cells 12 inside the chamber 18.

Like the first embodiment, the heating tube 14 can be made of a porous or dense material that can withstand SOFC operating conditions, such as a ceramic, high temperature metal, metal alloy, cermets, or a high temperature metal or metal alloy mesh. To enhance catalytic burning, both the inner surface of the casing 16 and the outer surface of the heating tube 14 are coated with catalytic material.

Optionally, the heating chamber 20 is filled with a solid-state porous foam matrix (not shown) that is able to withstand SOFC operating conditions. The

pores of the foam can be coated with catalytic material to further enhance catalytic burning therein.

Also optionally, and now referring to Figures 9(a) and (b), the fuel cells 12 can be embedded in a solid-state porous foam matrix 24 that has sufficient mechanical strength to support the fuel cells 12 in the system 10. The foam matrix 24 can be made of a material that is electronically conductive, in which case the foam matrix 24 acts as a current collector and also can act as a catalyst support for the cathode catalyst in each fuel cell 12.

Referring now to Figures 10(a) and (b) and 11 and according to another embodiment of the invention, the fuel cells 12 are divided into electrically isolated groups of "sub-stacks" 28. Longitudinally-extending planar partitions 30 are used to divide the fuel cells 12, and are made of a material that is able to withstand SOFC operating conditions. Such materials include ceramics such as SiC, Al_2O_3, SiO_2, MgO, ZrO_2, high temperature metals or metal alloys, cermets, ceramics coating with metals or metals coating with ceramic. When an electrically conductive metal is used in the partitions 30, the metal can be coated with an electrically insulating material to prevent shorting.

One longitudinal edge ("inside edge") of each partition 30 is attached to the outer surface of a longitudinally extending air distribution tube 32. The other longitudinal edge of each partition 30 extends close to the inner surface of the heating tube 14. The partition wall 30 can be perforated or dense.

The air distribution tube 32 has a plurality of longitudinally spaced perforations 34 that discharge air from the air distribution tube 32 and to the cathode of each fuel cell 12. Air is supplied into the bottom of the tube 32 from an air source (not shown) and flows upwards and out of each perforation 34. In order for air to be discharged at a relatively uniform pressure along the length of the tube 32, the perforations 34 increase in diameter upwards along the tube 32,

to compensate for a decreasing air pressure upwards along the tube 32. Alternatively or in addition, the air distribution tube wall can be sufficiently porous to allow the passage of air therethrough.

In operation, the partitions 30 serve to electrically isolate each fuel cell sub-stack 28 from another, but allows the flow of air between the sub-stacks 28. This electrical isolation enables the sub-stacks 28 to be electrically connected in series. Current is collected from the ends of each fuel cell 12 in the sub-stacks 28.

Alternatively, the partitions 30 can be electrically conductive such that all of the sub-stacks 28 are electrically connected in parallel. Also alternatively, the partitions 30 can be provided without the air distribution tube 32, in which case the inside edges of the partitions 30 extend inwards to contact each other.

Instead of or in addition to supplying air to the cathodes, the air distribution tube 32 can be used to heat reactant air and the fuel cells 12, by burning heating fuel inside the air distribution tube 32 to produce heat. By supplying heating fuel under sufficient pressure, some of the fuel will permeate through the air distribution tube 32 and burn outside the air distribution tube 32 and inside the oxidant flow chamber 22. Heat can also be supplied to the fuel cells 12 by burning fuel in the heating chamber 22 between the heating tube 14 and casing 16, as described in the second embodiment.

According to another embodiment of the invention and referring to Figures 12(a) and (b), an inner tubular combustor 15 is coaxially mounted inside the tubular air distribution tube 32. The combustor 15 is fluidly coupled to respective fuel and air sources and operates in the same manner as described above. As the tubular combustor 15 is spaced from the air distribution tube 32, an annular air flow channel is formed therebetween through which supply air or exhaust air can be flowed. This type of arrangement is expected to enhance heat transfer

from the combustor 15 to the fuel cell stack 12 and also enable uniform air distribution within the stack 12. Optionally, the air distribution tube 32 can have multiple combustor tubes (not shown)to improve the heat transfer to the annular air flow channel. The heating tube 14 can be present to provide additional heat to the stack 12, as shown in Figure 12(a), or omitted, as shown in Figure 12(b).

According to another embodiment and referring to Figures 13(a)-(d), the fuel cell system 10 can have a rectangular cross-sectional shape, which is particularly useful for portable applications like laptop computers. The system 10 has a combustor 15 enclosed within an air distribution tube 32 – the arrows in Figures 13(a)-(d) indicate air flow. The combustor 15 can have various designs: in Figure 13(a), the combustor 15 is an elongated rectangular structure; in Figure 13(b), the combustor 15 comprises multiple longitudinally extending heating tubes; in Figure 13(c), the combustor 15 comprises multiple longitudinally extending heating tubes surrounding a longitudinally extending air inlet tube 31 that serves to enhance heat transfer between the combustor 15 and the air. In Figure 13(d), the fuel cells 12 are electrically isolated into a plurality of substacks, and the combustor is the elongated rectangular structure as shown in Figure 13(a).

While the present invention has been described herein by the preferred embodiments, it will be understood to those skilled in the art that various changes may be made and added to the invention. The changes and alternatives are considered within the spirit and scope of the present invention.

What is claimed is:

1. A solid oxide fuel cell system comprising

(a) at least one longitudinally extending tubular solid oxide fuel cell comprising a ceramic solid state electrolyte layer and inner and outer electrode layers concentrically arranged around and sandwiching the electrolyte layer;

(b) a longitudinally extending heater mounted in thermal proximity to the fuel cell for providing heat to the fuel cell during start up and during operation as needed; and

(c) a longitudinally extending tubular thermal casing the inside of which defines a first reactant chamber for containing a first reactant, the fuel cell and heater;

wherein the outer electrode layer is fluidly communicable with the first reactant, and the inner electrode layer is fluidly isolated from the first reactant and fluidly communicable with a second reactant, such as fuel.

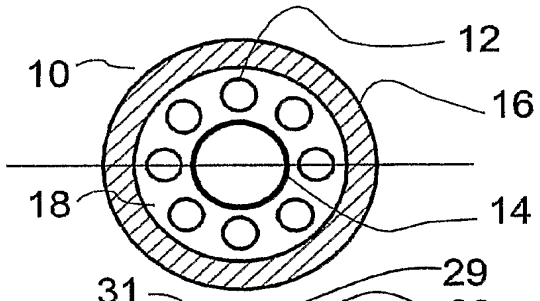

Fig. 1(a)

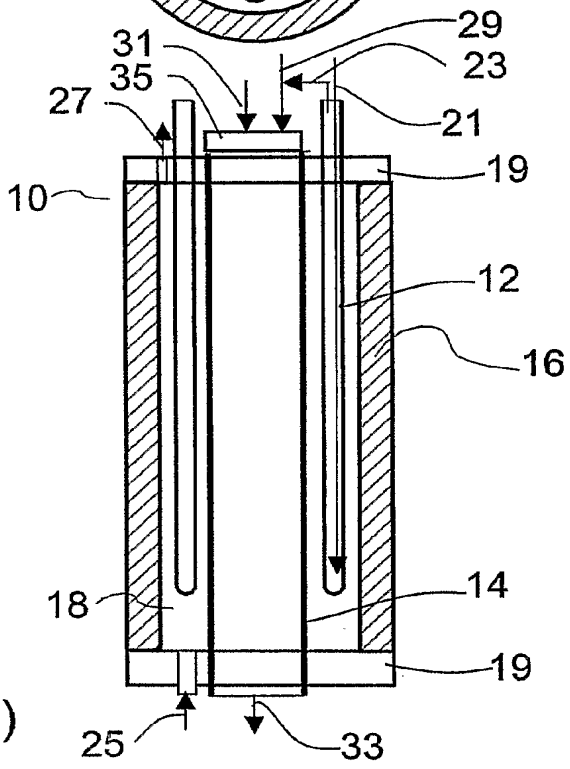

Fig. 1(b)

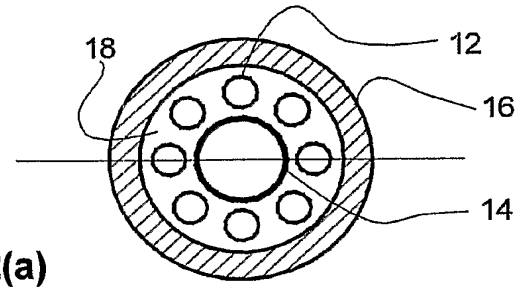

Fig. 2(a)

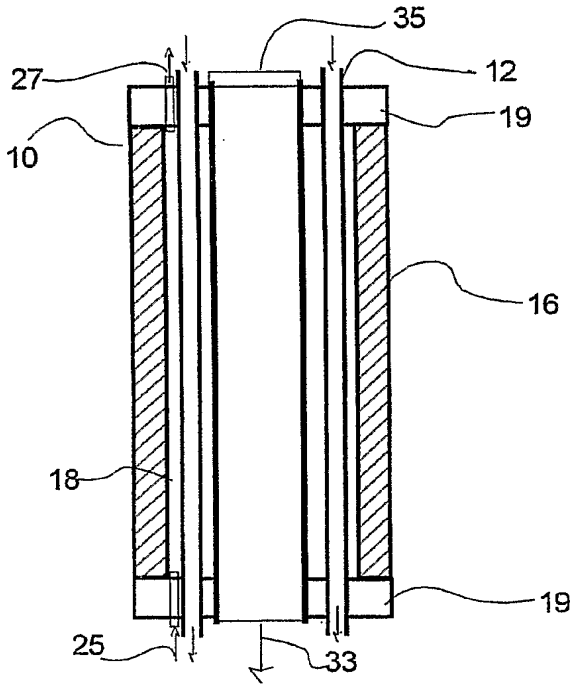

Fig. 2(b)

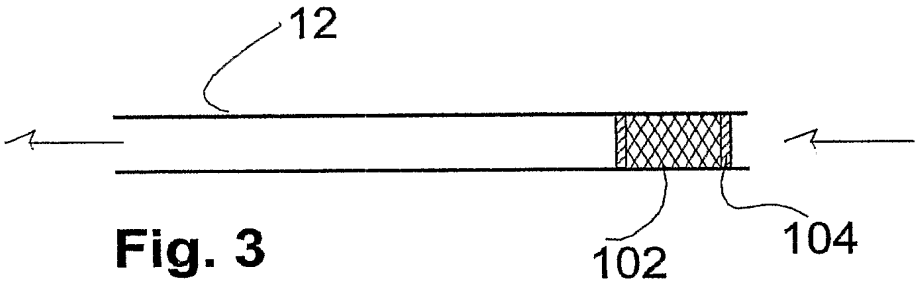

Fig. 3

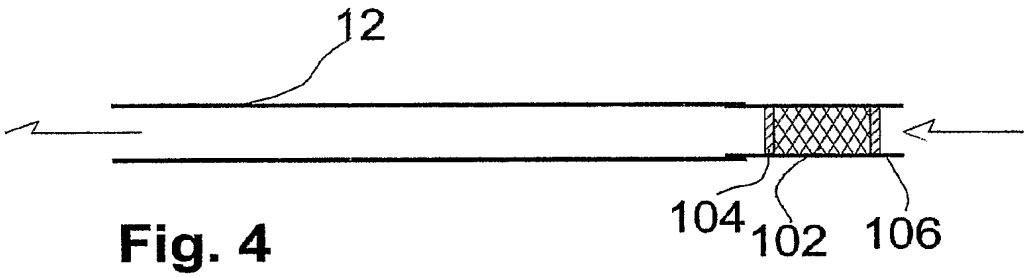

Fig. 4

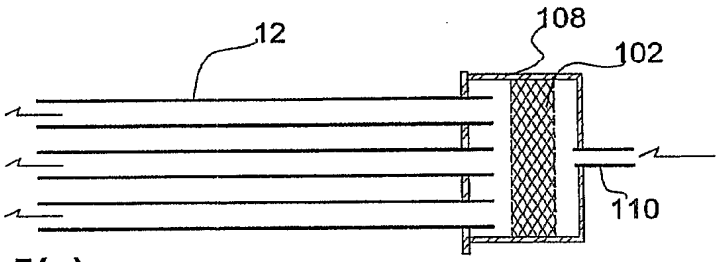

Fig. 5(a)

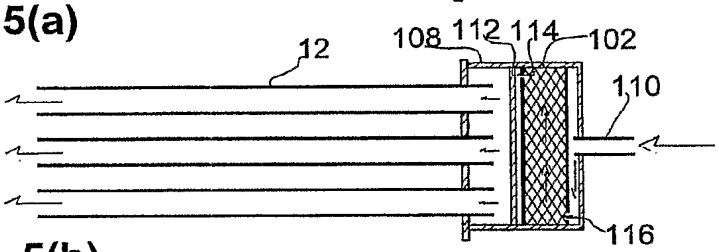

Fig. 5(b)

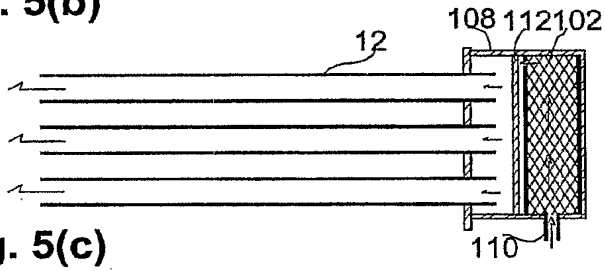

Fig. 5(c)

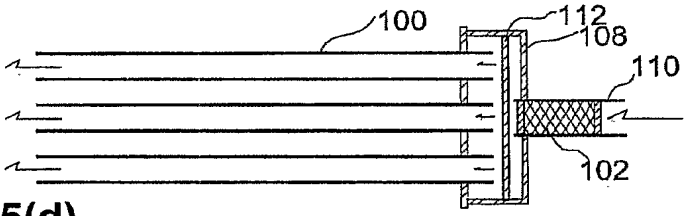

Fig.5(d)

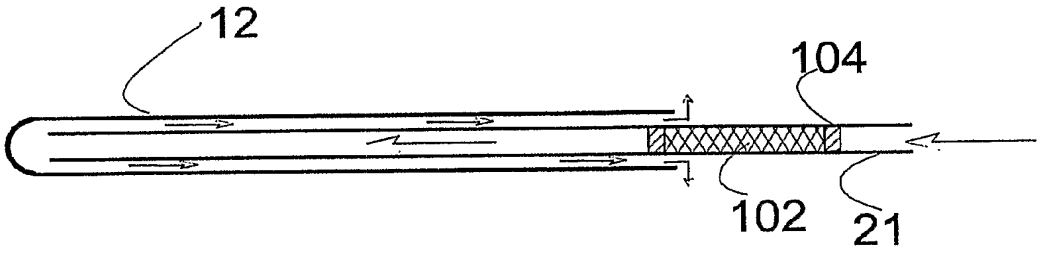

Fig. 6

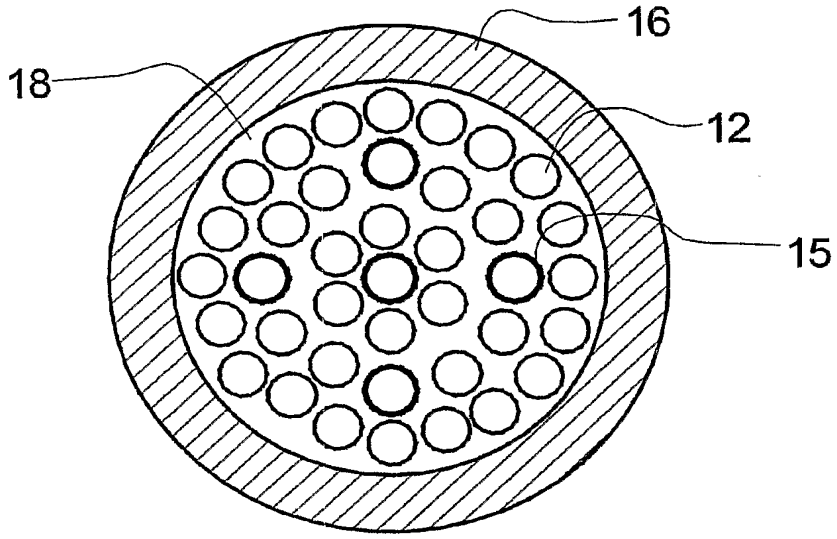

Fig. 7

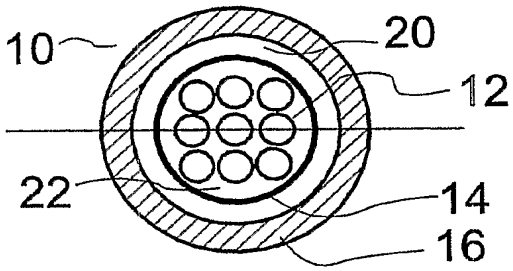

Fig. 8(a)

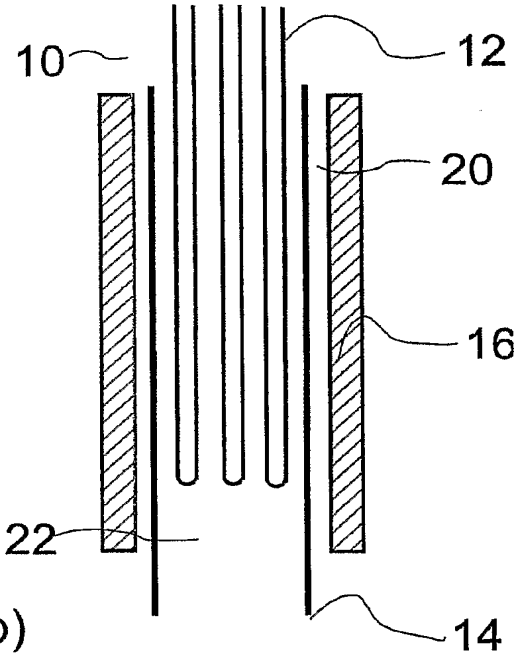

Fig. 8(b)

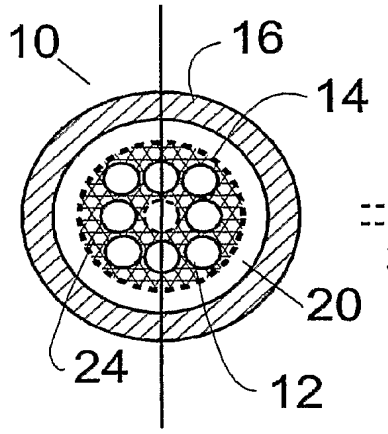

FIG. 9(a)

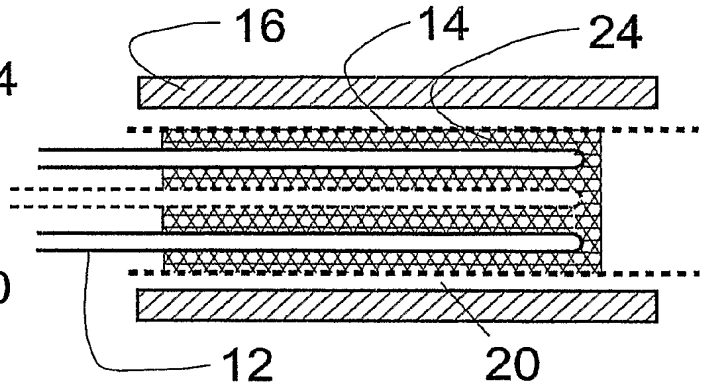

FIG. 9(b)

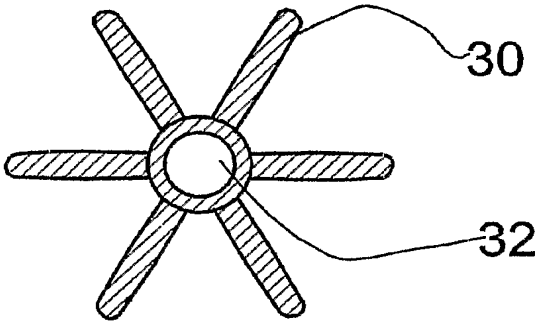

Fig. 10(a)

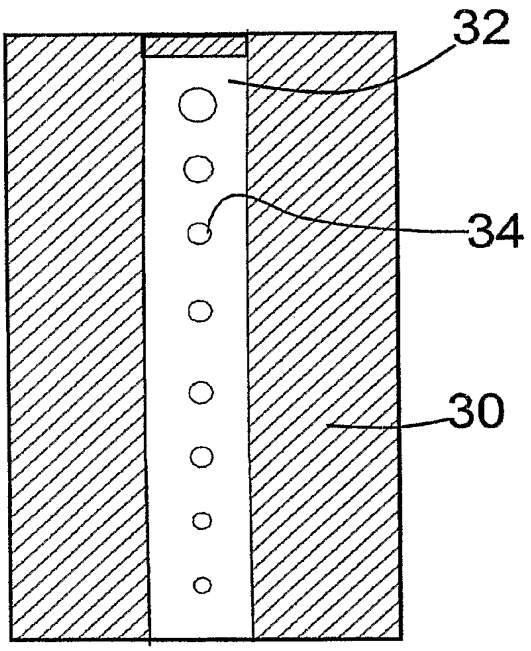

FIG. 10(b)

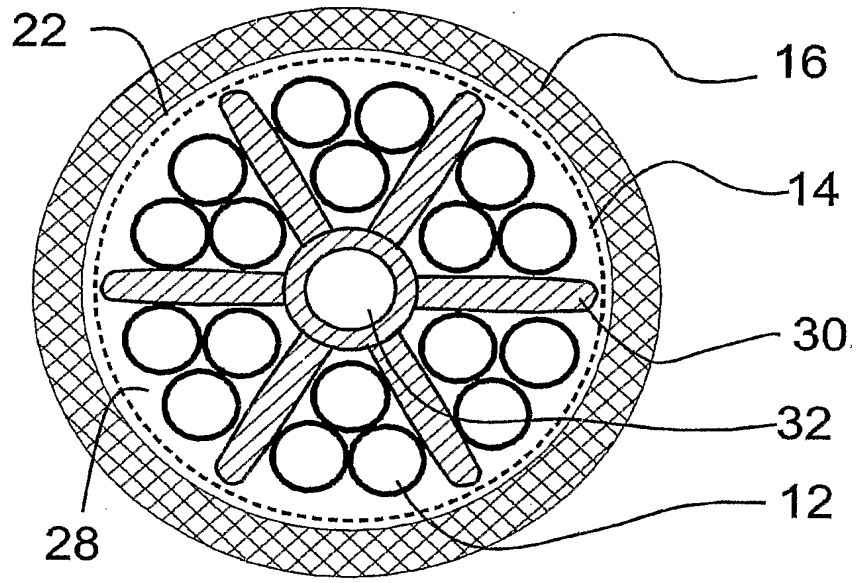

FIG. 11

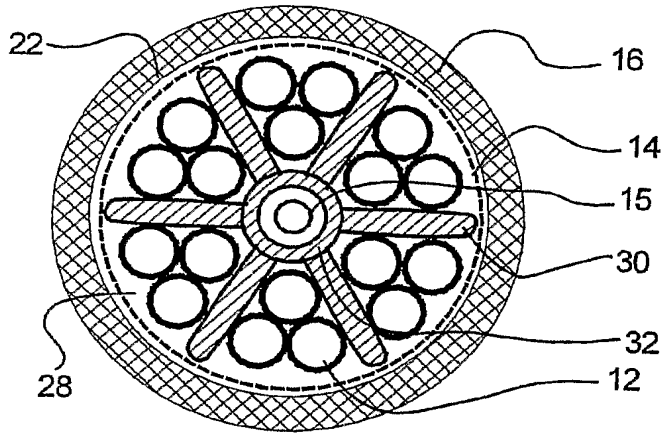

Fig. 12(a)

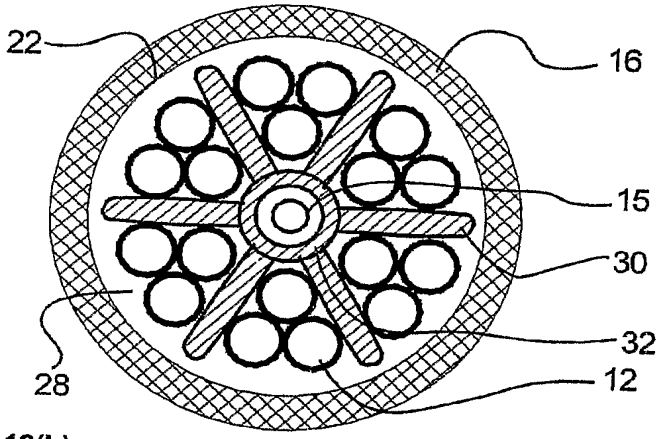

Fig. 12(b)

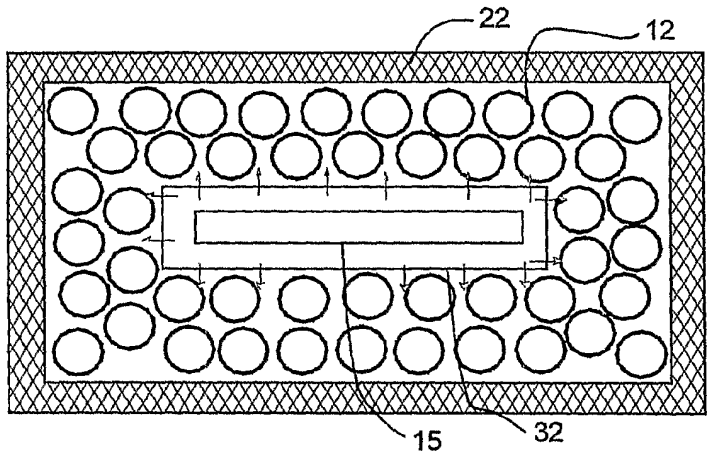

Fig. 13(a)

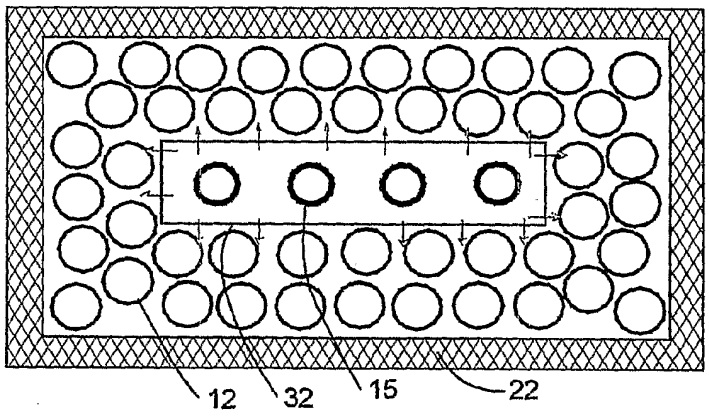

Fig. 13(b)

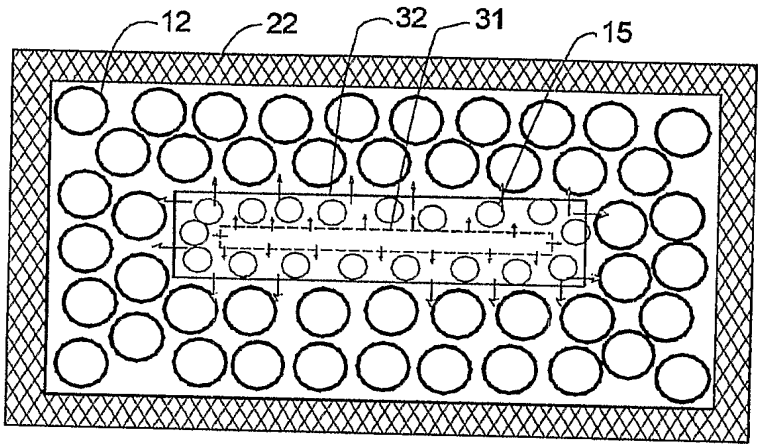

Fig. 13(c)

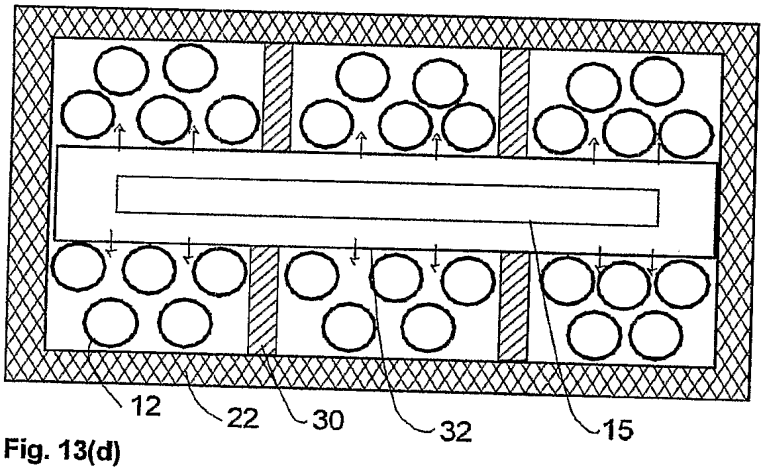

Fig. 13(d)